UCRL-JC-118385
PREPRINT

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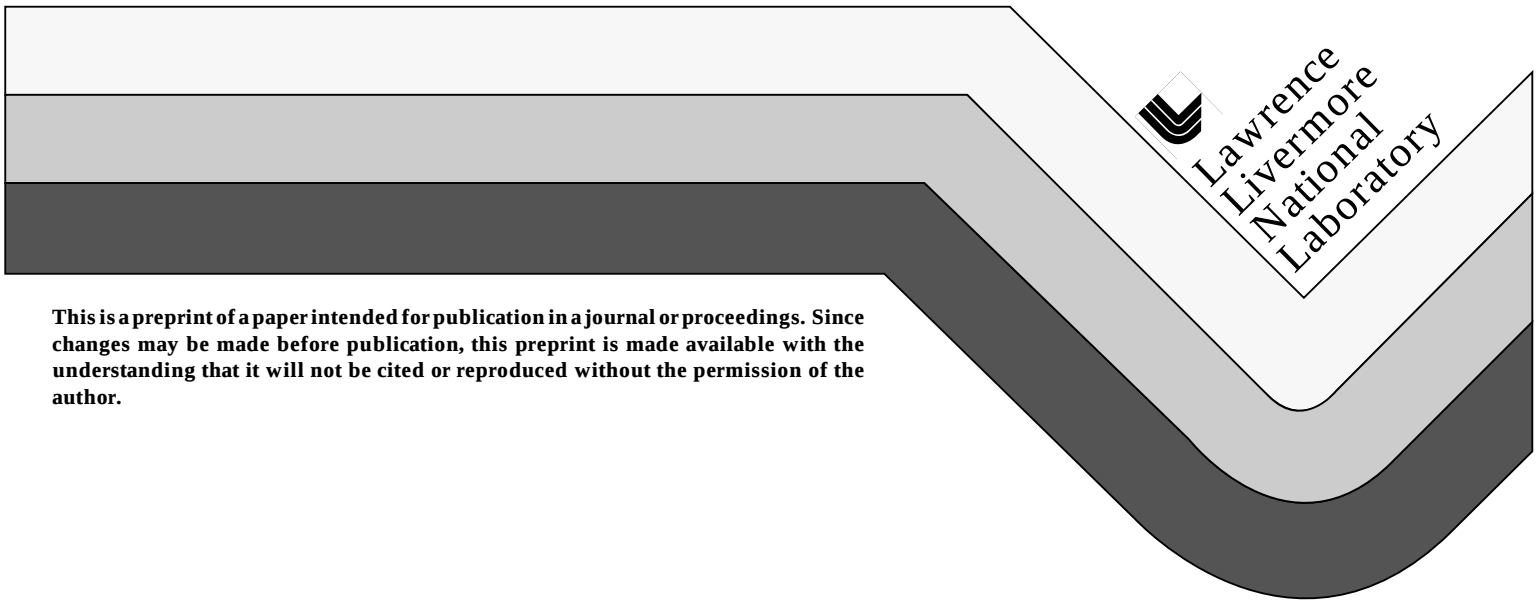
This paper was prepared for submittal to the
DOE Spent Nuclear Fuel-Challenges and Initiatives Conference
Salt Lake City, Utah
December 13-16, 1994

September 1994



QUALIFYING RADIOACTIVE WASTE FORMS FOR GEOLOGIC DISPOSAL

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ABSTRACT

We have developed a phased strategy that defines specific program-management activities and critical documentation for producing radioactive waste forms, from pyrochemical processing of spent nuclear fuel, that will be acceptable for geologic disposal by the U.S. Department of Energy. The documentation of these waste forms begins with the decision to develop the pyroprocessing technology for spent fuel conditioning and ends with production of the last waste form for disposal. The need for this strategy is underscored by the fact that existing written guidance for establishing the acceptability for disposal of radioactive waste is largely limited to borosilicate glass forms generated from the treatment of aqueous reprocessing wastes. The existing guidance documents do not provide specific requirements and criteria for nonstandard waste forms such as those generated from pyrochemical processing operations.

I. INTRODUCTION

Radioactive high-level waste (HLW) streams generated in the course of pyroprocessing of spent nuclear fuel must be solidified and packaged for eventual transfer to a licensed geologic repository. Before transfer, however, they must be approved for geologic disposal by the Department of Energy (DOE) Office of Civilian Radioactive Waste Management. Therefore, it is essential that the criteria for acceptability of HLW for disposal be clearly specified before solidification and packaging processes are fully developed. Because programs generating future HLW forms may span several years before actual production of waste, a documented program strategy and an implementation plan to guide HLW form development activities are critical.

The existing guidance, documentation, and DOE experience base for establishing the acceptability of HLW forms for geologic disposal is limited to borosilicate glass HLW generated at the Savannah River Site and West Valley Demonstration Project. In both cases, the HLW streams were generated from aqueous reprocessing operations, not pyrochemical processing, decades before the development and selection of borosilicate glass as the HLW form acceptable for geologic disposal. The existing documentation used to establish borosilicate glass as an acceptable HLW form cannot be used directly for HLW produced from pyrochemical processing operations, because of the fundamental differences in the nature of the waste streams involved.

The pyrochemical processing operations under consideration for treatment of DOE-owned and commercial-reactor spent fuel generate HLW streams that consist of (1) chloride salts of the fission products, (2) actinide elements in metallic form, and (3) cladding and/or fuel-element structural materials, also in metallic form. These streams require further treatment and consolidation before they can become acceptable HLW forms for geologic disposal. The strategy, planning, and critical documentation considered necessary for the successful development of acceptable pyroprocess HLW forms must be explicitly defined to guide process development. The development of the pyrochemical processing flowsheets can proceed concurrently with the development of possible solidified HLW forms. This allows some unique opportunities to be explored in changing the basic chemical compositions of HLW streams to arrive at an acceptable HLW form, an option not possible with the aqueous reprocessing wastes being dealt with at the Savannah River and West Valley sites.

We have developed a general strategy and framework for defining specific program-management activities and

the critical documentation to ensure that future consolidated HLW forms will be acceptable by the DOE for disposal. The required documentation spans several years, starting with the initial decision to develop a pyroprocessing technology for application to a particular spent-fuel class, and ending with the production of the last HLW form for geologic disposal. The strategy requires that an overall program schedule be developed that specifies the development schedule and the sequencing of these critical documents. These documents provide a focus for the development of pyrochemical processing and HLW form-production technologies. The documents provide the bases for the critical programmatic decisions that must be made to arrive at one acceptable HLW form for each major HLW stream requiring separate solidification and packaging for geologic disposal.

The general strategy described here for pyroprocessing technology is directly applicable to DOE programs that are also currently developing HLW forms for geologic disposal. These include the DOE Spent Nuclear Fuel Program, the Idaho National Engineering Laboratory program for high-sodium liquid wastes and calcine, and the Hanford program for the combined double-shell and single-shell tank HLW. The strategy is also applicable for future geologic disposal options being examined for disposition of excess weapons-grade plutonium by the newly created DOE Office of Excess Nuclear Materials.

II. MANAGING DISPOSAL OF HIGH-LEVEL WASTE

The Nuclear Waste Policy Act (NWPA) of 1982 assigned DOE the responsibility for managing the geologic disposal of spent fuel and HLW generated within the United States.[1] The NWPA defined a schedule and outlined a process for the DOE to follow. A 1985 Presidential Memorandum established that HLW from defense programs would be placed in the same geologic repository as commercial spent fuel and HLW. The Nuclear Waste Policy Act Amendments Act of 1987 designated a site at Yucca Mountain, Nevada, as the single site to be characterized for its suitability as a geologic repository.[2] The Amendments Act also requires DOE to report to the President and the Congress between 2007 and 2010 on the need for a second repository.

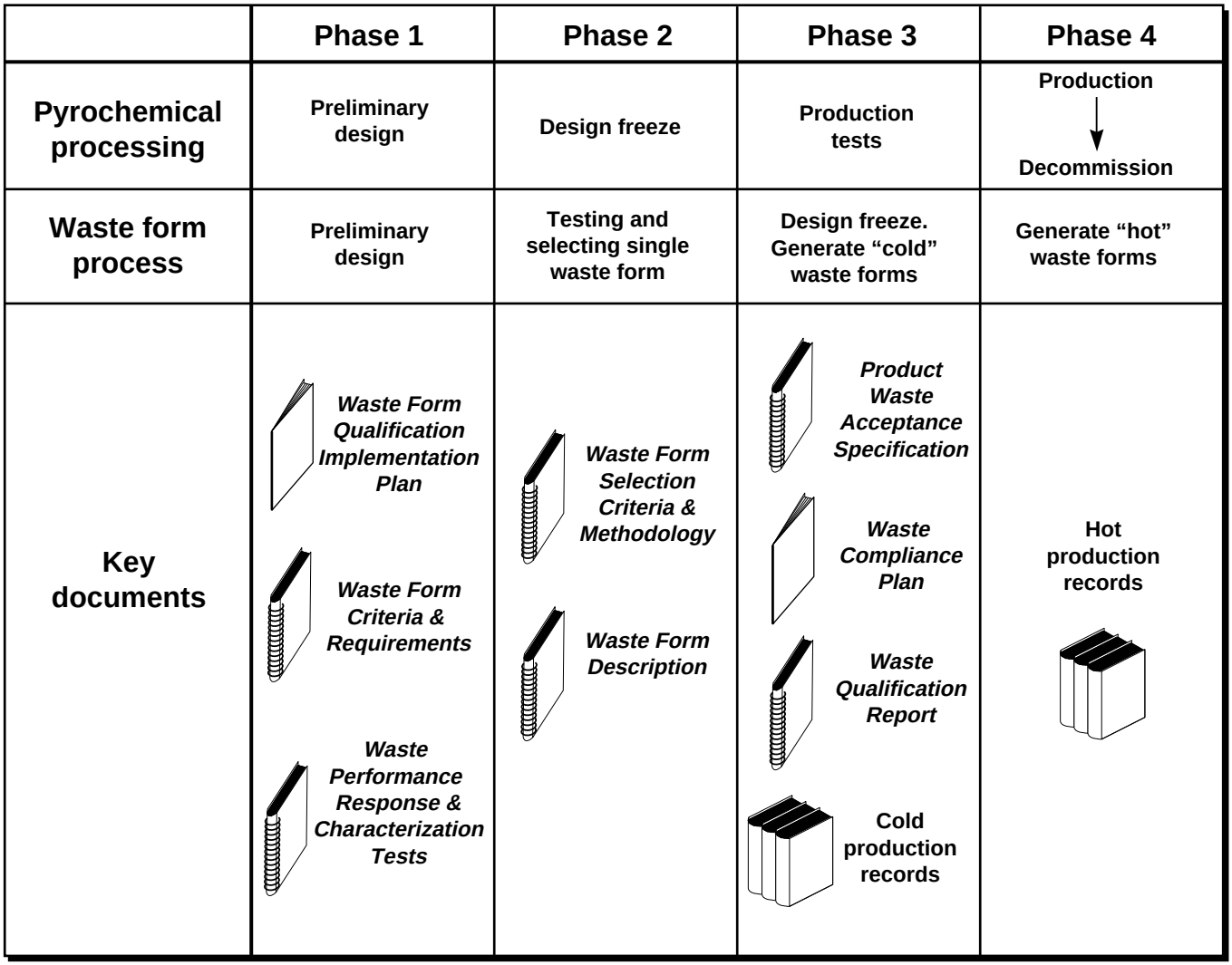
The NWPA defines spent fuel as fuel that has been withdrawn from a nuclear reactor following irradiation, the constituent elements of which have not been separated by reprocessing. Spent fuel includes the non-fuel components and hardware as identified in Title 10 of the *Code of Federal Regulations*, part 961. High-level waste is defined as the highly radioactive material resulting from the reprocessing of spent nuclear fuel. This includes the liquid waste produced directly in reprocessing, any solid materials derived from the liquid waste that contains fission products in sufficient concentrations, and other highly radioactive materials that have been determined by the Nuclear Regulatory Commission, consistent with the NWPA, to require permanent isolation. High-level waste can be either commercial or defense-related.

For many years prior to the passage of the NWPA, various programs within DOE were actively developing solidification processes and waste forms to stabilize HLW. Initial activities of the DOE Office of Defense Programs focused on the liquid wastes from the Defense Waste Processing Facility at the Savannah River Site, with later plans to address wastes at Hanford and the Idaho National Engineering Laboratory. Later, in parallel, DOE's Office of Nuclear Energy was developing a process and waste form to stabilize the liquid HLW from the West Valley Demonstration Project at the commercial spent-fuel reprocessing plant at West Valley, New York, as part of its remedial action efforts. In the early 1980s, borosilicate glass was selected as the single preferred encapsulation medium for both the Savannah River and West Valley sites.[3,4] In 1988, borosilicate glass was also selected for the Hanford double-shell tank HLW; a decision on treatment of the HLW in the Hanford single-shell tanks was deferred to a later date.[5]

It was recognized that the stabilized borosilicate HLW forms to be produced would be disposed of in a geologic repository, but that the schedules required to complete the HLW solidification processes at West Valley would result in the production of actual HLW years before the repository could be licensed and operated. As a result, DOE defined and implemented a formal waste acceptance process in 1985 to ensure that the HLW produced at these two facilities would be acceptable for disposal under the future repository license. This waste acceptance process has been documented in several places.[6,7]

For the Savannah River and West Valley sites, the top-level HLW requirements and criteria are defined in the *Civilian Radioactive Waste Management Systems Requirements Document* and the *Waste Acceptance System Requirements Document*.[8,9] Lower level, more detailed requirements and criteria are contained in the *Waste Product Specifications Document* for vitrified HLW.[10] The two top-level guidance documents currently are limited to standard borosilicate glass HLW and commercial spent fuel from light water reactors, and define the standard HLW form as borosilicate glass. Solidified HLW forms other than borosilicate glass that are likely to result from pyroprocessing HLW streams are defined as "nonstandard HLW."[9] However, these documents do not provide any specific guidance on the requirements and criteria for nonstandard HLW forms.

For future HLW producers whose waste form development of nonstandard HLW is under way and where the forms of the various HLW streams have not yet been selected, there is a special need to define the



requirements and criteria that must be satisfied to ensure that their future waste will be acceptable for disposal. These requirements must not be so restrictive as to create a premature constraint on possible process options. However, once a single preferred waste form such as glass or some other nonstandard form is selected, the requirements must be made very restrictive to focus and close the design and process development options.

III. PHASED DEVELOPMENT OF ACCEPTABLE WASTE FORMS

Four sequential periods or phases are used for defining and implementing a strategy to qualify for disposal HLW forms derived from pyroprocessing radioactive waste streams (Fig. 1). These phases start with the initial research and development activities, continue through the design and construction of both prototypical and full-scale HLW solidification and packaging plants, and end with the completion of the last production run of radioactive HLW forms. The time periods typically span several years, depending upon a specific pyrochemical process development and production operational schedule. Each succeeding time period is supported by the activities and documents generated in the preceding period. Therefore, the timing and scheduling of each period is critical to the successful completion of its successor period.

Figure 1. Four phases and key documents are required for qualifying geologic disposal of radioactive wastes from pyrochemical processing.

Phase 1 begins with the initial decision to develop a pyrochemical process that generates highly radioactive salt and metal waste streams. There is an opportunity during the initial flowsheet development of the

pyrochemical process to identify and incorporate major process modifications that facilitate the production of solid HLW forms from the radioactive waste streams. Probably the greatest opportunity to facilitate development of acceptable solid waste forms is to make changes to the main process chemical constituents. It is critical that the waste form development be started as soon as possible after initial design of the pyrochemical process, so that any waste process changes can be readily incorporated into development of the main process flowsheet.

During Phase 1, development of more than one solid waste form for each HLW stream is pursued to ensure that an acceptable waste form for geologic disposal can be developed in the subsequent periods. Phase 1 ends when the opportunity no longer exists to make further changes to the basic pyrochemical process flowsheet or to the processing materials and equipment. This generally occurs when the design configuration for the main pyrochemical process must be frozen so that the more detailed final design phase for the facility and equipment can be started. At this point, additional major changes to the main process configuration cannot be accommodated by design, equipment procurement, or construction schedules. In DOE design programs, the end of Phase 1 corresponds to the end of the Title 1 or preliminary engineering design phase activities.

Phase 2 starts immediately after Phase 1 ends. Because of the main process design freeze, the salt and metal waste stream compositions are now much more limited in their ranges than during Phase 1. Development of more than one solid HLW form for each waste stream may continue, however, to enhance the likelihood of successful development. The HLW form solidification development and design schedule generally lags that of the main pyrochemical process. Performance response testing and characterization measurements of the candidate solid waste forms are made in Phase 2 to guide waste form development. These tests and measurements include assessment of leach resistance, thermal stability, mechanical integrity, various physical properties, and resistance to deleterious radiolytic reactions. The results are documented and compared to the general requirements and criteria for waste forms destined for geologic disposal. The results from years of performance testing data and characterization measurements are used in selecting a single HLW form that is determined to be acceptable for geologic disposal and can be fabricated remotely on a production scale. The selection of a single waste form occurs at the end of Phase 2, when an adequate database is established or when the actual HLW form solidification process materials and equipment must be frozen for the continued design of the HLW solidification process. This typically occurs at the end of the Title 1 or preliminary design phase for the actual HLW form solidification process.

Phase 3 begins with the design freeze for the HLW solidification process and ends before the first full-scale radioactive, or "hot," HLW form is produced. During Phase 3, non-radioactive, or "cold," production runs of full-scale HLW forms are made. Additional, detailed performance response testing and characterization measurements of the single selected HLW form for each waste stream are continued, and the results are extensively documented. Phase 4 starts with the production of the first full-scale radioactive HLW form and ends when the last hot HLW form is produced and the decontamination and decommissioning phase of the facility is started.

The major programmatic milestones that occur within these four phases are used to develop a detailed plan and schedule for implementing a specific program strategy. The phases define different program activities to guide the development and implementation of the HLW solidification process for a specific pyrochemical process.

Several key documents are used to define and implement the strategy and programmatic framework over the four program phases. These are not the only documents, but the critical ones used to provide the basic programmatic framework. Documents used in the first two phases establish and document the technical bases for selecting a single HLW form for each waste stream as acceptable for disposal in a geologic repository. They also ensure that the more extensive HLW process design and operational phases can proceed with an acceptable level of programmatic risk. These and the remaining documents generated in Phase 3 provide the information needed for the licensing process and the start-up activities for HLW production.

A. Phase 1 Documentation

The *Waste Form Qualification Implementation Plan* specifies how the generic HLW qualification strategy is to be implemented over the four phases. It identifies the number of waste streams for which different waste forms must be developed. Program-specific milestones and schedules are developed to plan the implementation process. Key decision points are identified. Specific responsibilities are assigned to organizations for developing the key documents required by the HLW qualification strategy.

The *Waste Form Criteria and Requirements* document describes the minimum set of technical requirements and criteria for an HLW form that is proposed for geologic disposal as required by Title 10 of the *Code of Federal Regulations*, part 60. This document is used to provide guidance for the development of an HLW form that is acceptable for geologic disposal. The specifications for the criteria and requirements must not be overly constraining to the waste form development process, nor are they to be as detailed as those required after a single waste form is selected in Phase 2.

Determining the appropriate level of detail for the requirements and criteria is a major programmatic challenge. As the program proceeds, the document will be updated and level of detail increased appropriately.

The *Waste Form Performance Response and Characterization Tests* document outlines what performance response tests and characterization measurements will be used to generate data for evaluating the various HLW forms being developed. Details of the proposed test methods used and references to test procedures are contained in this document. The tests will include, but not be limited to, evaluation of groundwater leach resistance, thermal and radiation stability, and physical and mechanical properties of the proposed waste forms.

B. Phase 2 Documentation

The *Waste Form Selection Criteria and Methodology* document specifies criteria (in addition to those cited in *Waste Form Criteria and Requirements* of Phase 1) to screen and select a single HLW form for each different waste stream from among the multiple forms under development. This document will also describe the methodology or process to be used in selecting the single waste form for each HLW stream. The document should be developed early in Phase 2, before selection of the final single waste form is made, to avoid biasing the selection process.

The *Waste Form Description* document provides the information required by the *Waste Form Criteria and Requirements* document created in Phase 1 on the specific HLW forms to be considered for geologic disposal. It describes how well the consolidated HLW form satisfies the criteria and requirements and gives the performance response and characterization test results. The information on performance response and characterization tests must be detailed enough to allow the proposed waste forms to be accepted into the planning base for a future geologic repository. Release of preliminary drafts or progress issues of this document are recommended during Phase 2 so that external stakeholder reactions can be incorporated into the final version.

C. Phase 3 Documentation

The *Product Waste Acceptance Specification* (PWAS) document provides the more detailed specifications that apply for the specific selected HLW form to be accepted by DOE's Office of Civilian Radioactive Waste Management for geologic disposal. This document is more detailed than the *Waste Form Criteria and Requirements* document issued in Phase 1 and is specific to a single selected HLW form. Versions of this document have been generated for borosilicate glass HLW at the Savannah River and West Valley sites.[10]

The *Waste Compliance Plan* details how the HLW producer will accomplish compliance with the specifications in the PWAS and document it in a *Waste Qualification Report*.

The *Waste Qualification Report* contains a compilation of test results and analyses that confirm that a specific waste form, when produced in a controlled fashion, will comply with specifications in the PWAS. It is more detailed than the *Waste Form Description* document of Phase 2.

Cold production records provide information on the HLW product characteristics as obtained from actual full-scale production runs of simulated forms using non-radioactive materials. These records will verify that the HLW forms have been produced in a controlled fashion and will comply with the PWAS.

D. Phase 4 Documentation

Hot production records contain the same type of technical information as for the cold production runs, but give information using full-scale actual HLW production with fully radioactive waste streams. These records are used as part of the operating licensing requirements to ensure the acceptability of the waste forms for geologic disposal.

IV. CONCLUSIONS

A series of program life-cycle documents has been identified to provide a general programmatic strategy for developing HLW forms requiring geologic disposal. The strategy is being used to develop HLW forms from pyroprocessing operations that are acceptable to DOE and their regulators. These documents, their scheduling sequence, and major interdependencies have been summarized. The strategy and documents cover an entire program's life cycle, commencing with an initial decision to develop a pyrochemical process that generates radioactive HLW streams requiring solidification and geologic disposal and ends with production of the last HLW forms acceptable for disposal. The strategy requires that each specific program establish the schedule and responsibility for generating the proposed documents at the earliest opportunity, based on the overall program summary schedule.

Although developed for a DOE pyrochemical processing program, this strategy is directly applicable to other ongoing DOE programs. These programs include, but are not limited to, the geologic disposal of (1) special DOE and research spent fuels (DOE EM-37), (2) high-sodium liquid wastes and calcine at Idaho National Engineering Laboratory, and (3) excess plutonium from dismantled weapons. These DOE programs, and particularly DOE-RW, should consider this strategy as

they develop their own programmatic approaches for all radioactive wastes requiring geologic disposal.

ACKNOWLEDGMENTS

Work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-Eng-48 and Argonne National Laboratory under Contract W-31-109-Eng-38.

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